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Optical properties of terbium-oxide films*

TADEUSZ MARCINÓW, CECYLIA WESOŁOWSKA, KRYSTYNA ŻUKOWSKA

Institute of Physics, Technical University of Wrocław, Wybrzeże Wyspiańskiego 27, 50–370 Wrocław, Poland.

Results of the investigation of terbium-oxide film deposition are presented. Terbium oxide films have been obtained by thermal evaporation. Immediately after evaporation onto heated substrates (up to 200°C) TbO_x layers ($1.5 < x < 1.75$) are formed These layers have been subjected to the oxidation process in air at temperature of 500°C and then to reduction process in hydrogen at temperature of 700°C. Thereupon Tb_4O_7 and Tb_2O_3 films are formed. Optical properties of these layers have been investigated. The optical constants, $n = f(\lambda)$ and $k = f(\lambda)$, have been determined in the wavelength range of 200–2500 nm.

1. Introduction

Up to now, optical properties of terbium-oxide layers have not been investigated sufficiently. The refractive index for Tb_2O_3 layers at $\lambda = 700$ nm ($n = 1.93$) was given in paper [1]. The authors have stated that the layer of 170-nm optical thickness turned out to be nonabsorbing at $\lambda \geqslant 330$ nm. The refractive indices for Tb_2O_3 ($n = 1.92$) and Tb_4O_7 ($n = 1.94$) layers at $\lambda = 550$ nm were given in paper [2]. The dependence of absorption coefficient (α) on the relative oxygen ion content in $TbO_{(1.75-x)}$ layers for $0 < x < 0.25$ near the absorption edge was also studied [3]. The absorption of Tb_4O_7 in the visible region was found to be caused by electron transitions from valence band to f level of tetravalent terbium ions. Investigations of optical properties of terbium oxide layers, grown by thermal evaporation in vacuum and subject to additional annealing in air and next to reduction process in hydrogen atmosphere have been presented in this paper.

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2. Experiment

Terbium-oxide layers have been evaporated by an electron beam method onto quartz substrate under the following conditions: the pressure in the vacuum chamber of $p = 1 \times 10^{-5}$–8×10^{-5} Torr kept constant by an air dosage, the substrate temperature raised from the room temperature up to 200°C, evaporation rate being about 3 nm/min. Under these conditions TbO_x layers have been formed. These layers have been annealed in the air at temperatures ranging from 400°C to 500°C for several hours and next subject to reduction process. In order to obtain nonabsorbing layers the reduction process was carried out in the special quartz tube in the flowing hydrogen atmosphere at temperature of about 700°C. Time of the reduction process depends on the layer thickness being nearly 4 hours for layers about 400 nm thick. To reach the highest degree of reduction all the layers examined have been heated for 5 hours. The measurements of reflectivity (R) and transmittivity (T) for terbium-oxide films were made immediately after the evaporation, after the annealing in air and reduction process in hydrogen atmosphere. These measurements have been carried out with Zeiss spectrometers such as: SPECORD UV VIS, SPECORD 61 NiR and UR–10 with reflection attachments for near-normal incidence of light. Thickness of the layers, measured by the multibeam interference method, varied from 140 nm to 950 nm.

3. Results and discussion

Figure 1 shows exemplary transmission coefficient dependence on optical wavelength $T = f(\lambda)$ for the layer 303 nm thick evaporated onto the quartz substrate at temperature 200°C. The curve **a** refers to the as-evaporated layer which was reddish-brown. The curves **b** and **c** represent the same layer after annealing in the air atmosphere at temperature of 500°C and after reduction process in hydrogen atmosphere, respectively. It should be emphasized that an increase in annealing and reducing times does not affect changes of the spectral characteristics (curves **b** and **c**). In virtue of this statement it is possible to conclude that after annealing in the air the layer composition does not change and is near or equal to that of Tb_4O_7 layers, whereas after the reduction process in hydrogen atmosphere the obtained layers have the spectral characteristics $T = f(\lambda)$ similar to those of the other sesquioxides of the rare earths [4] including the layers of Tb_2O_3 type. The layers immediately after the evaporation are of TbO_x type, where $1.5 < x < 1.75$. The optical constants n and k for Tb_4O_7 and Tb_2O_3 layers have been determined from $R = f(\lambda)$, $T = f(\lambda)$ characteristics, and the layer thickness d has been evaluated in nonabsorbing regions from the interference maximum positions or in the absorbing region by methods of Vriens and Rippens [5] and of Schulz and Tangherlini [6]. Results of the

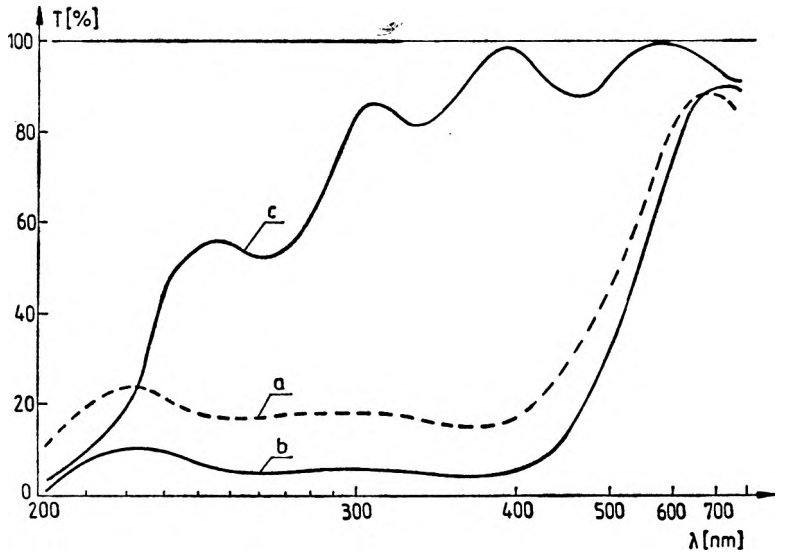

Fig. 1. Transmission coefficient T of terbium oxide layers: a — after evaporation, b — after annealing in air, c — after reduction in hydrogen

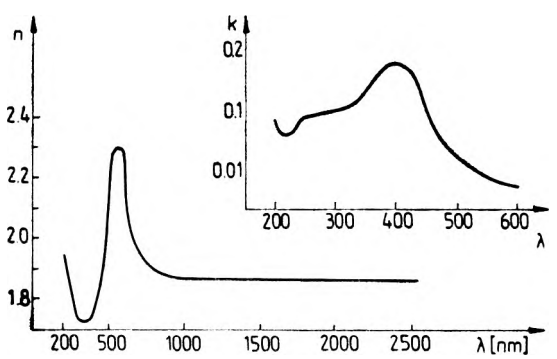

Fig. 2. Optical constants n and k for Tb_4O_7 layers

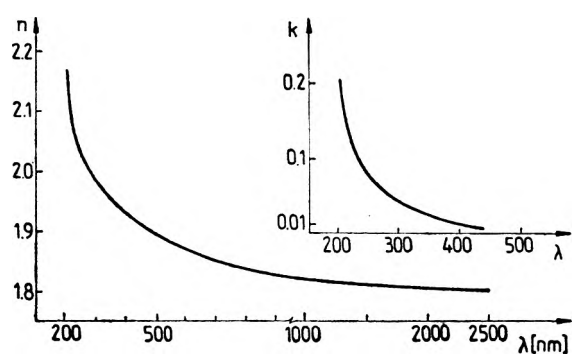

Fig. 3. Optical constants n and k for Tb_2O_3 layers

calculation of $n = f(\lambda)$ and $k = f(\lambda)$ for Tb_4O_7 and Tb_2O_3 layers are presented in Fig. 2 and Fig. 3, respectively.

Based on these investigations it can be stated that the refractive index of Tb_4O_7 exhibits a small dispersion in the near-infrared region (800–2500 nm) and for $\lambda = 1000$ nm equals 1.87. The anomaly of refractive index occurs in visible region with the maximum at $\lambda = 550$ nm. An increase in k value for the wavelengths shorter than 600 nm is connected with the presence of the absorption band. In the wavelength ranging from 250 nm to 410 nm the absorption coefficient $\alpha = 4\pi k/\lambda$ is nearly constant and equals 0.75×10^5 cm^{-1} thus being close to the value given in [3]. The region of good transparency ($\alpha \leqslant 10^3$ cm^{-1}) spreads from 640 nm towards the infrared. The refractive index for Tb_2O_3 layers is slightly lower in the infrared region than that for Tb_4O_7 layers at $\lambda = 1000$ nm, and n equals 1.84, for $\lambda = 550$ nm, $n = 1.85$. The refractive index values obtained for Tb_2O_3 and Tb_4O_7 layers are lower than those given in [1, 2]. As shown in Fig. 3 the normal dispersion of the refractive index for Tb_2O_3 layers was found in the region examined (200–2500 nm), the refractive index slightly changes in the infrared region while in ultraviolet Tb_2O_3 layers exhibit weak dispersion. The region of a good transparency spreads from $\lambda = 390$ nm. Beyond that region like other rare earth oxides [4], Tb_2O_3 as well as Tb_4O_7 show a narrow absorption band in near infrared (~ 2.7 μm) due to the presence of the absorbed hydroxyl group (OH).

4. Conclusions

Investigations of optical properties of terbium-oxide layers have led to the following conclusions:

i) there is a possibility of modifications of optical properties of terbium-oxide layres,

ii) Tb_2O_3 layers can be obtained by the process of reduction of Tb_4O_7 layers in hydrogen at high temperatures.

The Tb_2O_3 layers might be used for optical purposes in the 400–2500 nm region, but — as it can be seen — the layer deposition technology is difficult due to necessity of carrying the reduction process in hydrogen atmosphere. On the other hand, Tb_4O_7 layers might be used as the filters cutting down the visible and ultraviolet ranges of spectrum or in multilayer systems in near-infrared region $\lambda > 640$ nm. All layers examined exhibit very good chemical and mechanical resistance.

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Оптические свойства слоев окиси тербия

Представлены результаты исследований получения слоев окиси тербия. Сразу же после испарения на подогретом основании (до $T = 200°C$) были получены слои TbO_x ($1{,}5 < x < 1{,}75$) которые затем подвергали процессам окисления в воздухе при температуре 500°C и восстановления водородом при температуре 700°C – в результате чего возникли слои Tb_4O_7 и Tb_2O_3. Были исследованы оптические свойства этих слоев, а также определены их оптические постоянные: $n = f(\lambda)$ и $k = f(\lambda)$ для волн длиной в 200–2500 нм.